

# Scaling relation of spin wave lifetime in double-exchange systems

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Spin excitation spectrum of CMR manganites are studied from both theoretical and experimental sides. Scaling relations for the spin wave lifetime in the electronic model with double-exchange interaction is observed, which is consistent with results for neutron inelastic scattering experiments for  $(\text{La,Y})_{0.8}\text{Sr}_{0.2}\text{MnO}_3$ . Roles of other interactions and degrees of freedom such as polaron effects are also discussed.

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## I. INTRODUCTION

One of the main issues in colossal magnetoresistance (CMR) manganites  $(\text{La,A})\text{MnO}_3$  is to determine the appropriate model to understand the mechanism of its magneto-transport phenomena. Canonical model for these compounds is the double-exchange (DE) Hamiltonian introduced by Zener [1] in order to study the ferromagnetism. Transport properties in DE systems are recently studied in a controlled manner using the dynamical mean-field approach by one of the authors [2].

Another candidate model for CMR manganites is the system which incorporates large electron-lattice couplings [3,4]. Effects of Anderson localization in the DE systems with and without charge disorder [5–7] are also investigated. Roles of orbital degeneracies in  $e_g$  electrons have also been studied in order to explain several transport properties [8,9].

One way to justify the relevance of the model is to compare universal behaviors insensitive to parametrization, such as scaling relations, with experimental results. For example, in compounds with relatively wide bandwidth  $W$  and high Curie temperature  $T_c$  such as  $(\text{La,Sr})\text{MnO}_3$  with sufficient doping concentration, it has been shown that magnetoresistance satisfy the scaling relation [10]

$$-\Delta\rho/\rho_0 \propto M^{*2} \quad (1.1)$$

if not affected by extrinsic effects such as grain boundary effects [11]. Here,  $\rho_0$  is the resistivity at the paramagnetic phase in the absence of magnetic field,  $\Delta\rho$  is the change in resistivity under magnetic field, and  $M^*$  is the magnetization normalized by its saturation value in the form  $M^* = M/M(T \rightarrow 0)$ . From theoretical point of view, this result is reproduced by the calculation within DE

Hamiltonian described above [2], which shows that this model is possibly a relevant model to describe magneto-transport properties of these compounds at least in these scaling regime.

On the other hand, compounds with relatively narrow bandwidth  $W$  and low  $T_c$  show several different behaviors on magnetic and transport properties. For example, scaling relation for resistivity and magnetism which differs from eq. (1.1) are reported for thin film data in  $(\text{La,Ca})\text{MnO}_3$  [12].

Under the assumption of strong electron-lattice effect, semiconductive behavior above  $T_c$  typically observed in these narrow  $W$  compounds is explained by formation of lattice polaron with dynamic JT distortion [3]. In these compounds, large lattice distortions in oxygen are indeed observed in  $(\text{La,Ca})\text{MnO}_3$  [13–15]. However, there are no reports so far which relates theory with scaling relations in experiments for low  $T_c$  compounds. Therefore it is not clear at this moment whether lattice effect is relevant or not to universalities in magneto-transport properties of narrow  $W$  (low  $T_c$ ) compounds.

Investigation of spin excitations is another approach to study the nature of CMR manganites, since the ferromagnetism in these systems reflects the nature of electronic states [16]. Experimentally, spin excitation spectrum of  $(\text{La,A})\text{MnO}_3$  ( $A=\text{Pb, Sr, Ca}$ ) has been measured by neutron inelastic scattering measurements [17–21]. In  $\text{La}_{0.7}\text{Pb}_{0.3}\text{MnO}_3$  which is classified as a wide  $W$  compound with ferromagnetic metal phase at low temperature, spin wave dispersion in cosine-band form is observed throughout the Brillouin zone.

Theoretically, it is shown that double-exchange Hamiltonian on a cubic lattice reproduces cosine-type dispersion relation

$$\omega_q \propto 3 - (\cos q_x + \cos q_y + \cos q_z) \quad (1.2)$$

in the strong Hund's coupling limit within the spin wave approximation, *i.e.* lowest order  $1/S$  expansion. In Fig. 1 we show the comparison between the experiment [18] and theory [22]. We see that the DE system also accounts for the spin wave excitation in CMR manganites. From the fitting, we obtain the values for electron hopping energy  $t$  and Hund's coupling  $J_H$ . These parameters are also satisfactory in comparison with experiments in the way that  $t$  is consistent with the estimate for  $e_g$  electron bandwidth

and also in the way that the estimate for  $T_c$  for the DE model is consistent with experimental value [22].

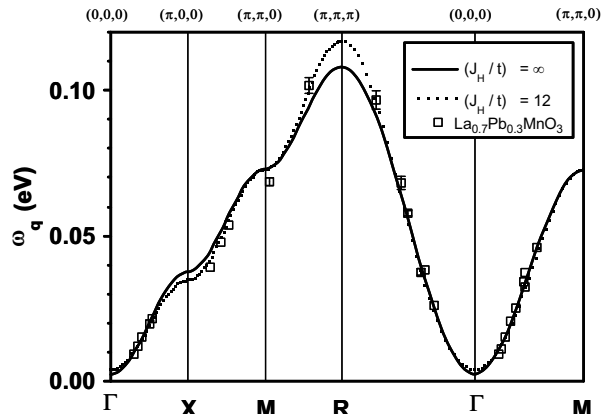


FIG. 1. Spin wave dispersion relation for the DE Hamiltonian for  $J_H/t = \infty$  (solid curve) and  $J_H/t = 12$  (dotted curve). Experimental data (squares) are taken from ref. [18].

For the case of finite  $S$ , exact diagonalization studies have been performed [23,24]. Quantum fluctuations at finite  $S$  produces deviation from the cosine band, nevertheless the deviation is small if doping concentration is large enough [24]. Therefore, as long as the dispersion relation is concerned, DE Hamiltonian also accounts for the spin excitation of CMR manganites with wide  $W$ .

Anomalous temperature dependence in spin wave lifetime is also observed in  $\text{La}_{0.7}\text{Pb}_{0.3}\text{MnO}_3$  [18]. Line width of spin wave substantially expanded by increasing temperature. This is typically observed at zone boundary, *i.e.* high energy part of the spin wave dispersion relation. Since the spin wave dissipation is determined by the presence of excitations coupled to spins, study of spin wave lifetime clarifies not only the spin excitation behavior but also the existence of excitation spectrum for other degrees of freedom.

In this paper we investigate the lifetime of the spin wave excitations from both theoretical and experimental sides. For theoretical approach, we study DE system, while experimentally we study several compounds with relatively wide bandwidth. We aim to obtain scaling relations in spin wave lifetime and establish possible relevance between theory and experiment.

## II. THEORETICAL AND EXPERIMENTAL APPROACHES AND RESULTS

From the theoretical side, we study the Kondo lattice model with ferromagnetic spin exchange,

$$\mathcal{H} = -t \sum_{\langle ij \rangle, \sigma} \left( c_{i\sigma}^\dagger c_{j\sigma} + h.c. \right) - J_H \sum_i \vec{S}_i \cdot \vec{\sigma}_i, \quad (2.1)$$

in the strong Hund's coupling region  $J_H/t \gg 1$ . We consider the system on a cubic lattice with nearest neighbor

hoppings. We investigate the spin wave lifetime in relation with magnetic moment, and clarify the role of DE mechanism with respect to dissipation of spin excitations.

Green's function  $G_\sigma(k, \omega) = (\omega - \varepsilon_k - \Sigma_\sigma(\omega))^{-1}$  at finite temperature is obtained from the dynamical mean-field approximation [25] on a cubic lattice. Here,  $\Sigma_\sigma(\omega)$  is a  $k$ -independent self energy. Using these Green's functions, Spin susceptibility  $\chi(q, \omega)$  are calculated through electron polarization diagrams. Within the lowest order of  $1/S$  spin wave expansion [22], real part of the self-energy for spin wave determines the dispersion relation through the self-consistency equation

$$\omega_q = \text{Re} \Pi(q, \omega_q), \quad (2.2)$$

and the self-energy is constructed from the spin polarization diagrams for fermions and vertices proportional to  $J_H$ . Spin wave line width  $\Gamma_q$  is obtained by the imaginary part of the self-energy,

$$\Gamma_q = \text{Im} \Pi(q, \omega_q) \propto \text{Im} \chi(q, \omega_q). \quad (2.3)$$

Physical interpretation of this result is that the origin of the spin wave line width is the dissipation process of spin wave into spin excitations in Stoner continuum.

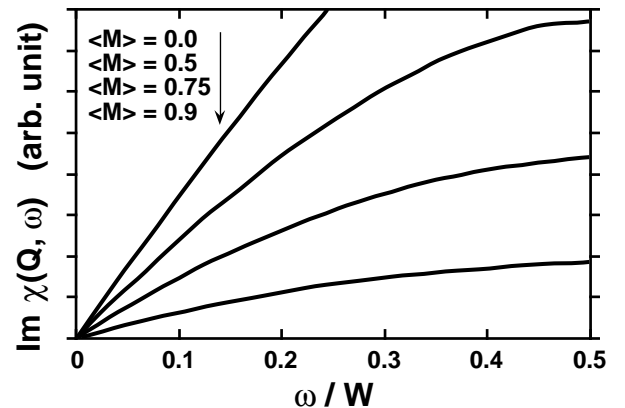


FIG. 2. Imaginary part of the Stoner susceptibility (Stoner absorption)  $\chi(Q, \omega)$  at the zone boundary  $Q = (\pi, \pi, \pi)$  for various temperatures.

In Fig. 2 we show  $\text{Im} \chi(Q, \omega)$  at the Brillouin zone boundary  $Q = (\pi, \pi, \pi)$  for various temperatures, where we take  $J_H/W = 2$  and  $x = 0.3$ . Here  $W = 6t$  is the electron bandwidth. We see that at small  $\omega$  we have  $\omega$ -linear relation, *i.e.*  $\text{Im} \chi \propto \omega$  at  $\omega \ll W$ . Coefficients for  $\omega$ -linear part decrease by decreasing the temperature. In Fig. 3 we show the coefficient for  $\omega$ -linear part  $\partial \text{Im} \chi(Q, \omega) / \partial \omega|_{\omega \rightarrow 0}$  as a function of normalized magnetization  $M^*$  at wave vector  $Q$ . As a result we find

$$\text{Im} \chi(Q, \omega) \propto (1 - M^{*2}) \omega \quad (2.4)$$

for small values of  $\omega$ . The relation (2.4) is observed at all values of  $q$  with slightly different coefficients. Since  $\Gamma_q \propto \text{Im} \chi(q, \omega_q)$ , we obtain the relation

$$\Gamma_q = \alpha_q(1 - M^{*2})\omega_q \quad (2.5)$$

where  $\alpha_q$  is a dimensionless function weakly dependent on  $q$ .

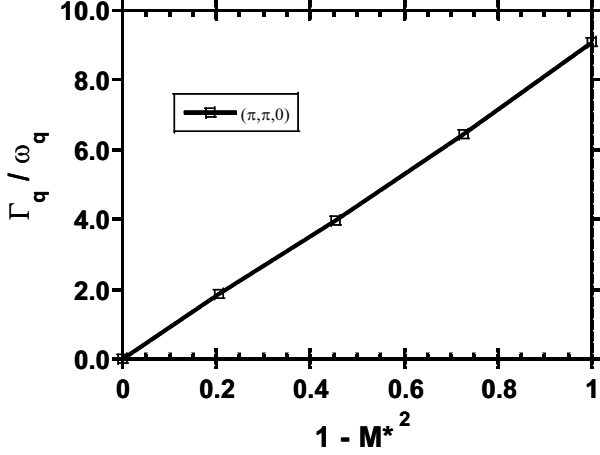


FIG. 3. Theoretical results for scaling behaviors in inverse lifetime of the spin wave in the form  $\Gamma_q/\omega_q \propto 1 - M^{*2}$ .

Now, we show experimental results for spin wave lifetime with respect to relatively wide  $W$  (high  $T_C$ ) samples of  $((\text{La}, \text{Y}), \text{Sr})\text{MnO}_3$ .

Single crystals of  $(\text{La}_{1-x-y}\text{Y}_y)\text{Sr}_x\text{MnO}_3$  (typically  $6\phi \times 80$  mm) were grown by the floating-zone method. The end part (30–40 mm) were used for neutron measurements. In order to study the effects of electronic bandwidth, three samples were chosen;  $y = 0.00, 0.05$  and  $0.10$  with the Sr concentration fixed at  $x = 0.20$ . The tolerance factor  $t$ , which is defined as  $t = (r_A + r_O)/\sqrt{2}(r_B + r_O)$  for  $\text{ABO}_3$  perovskites and scales with the band width, decreases with doping Y; 0.908, 0.906 and 0.903 for  $y = 0.00, 0.05$  and  $0.10$ , respectively. These values are, however, still closer to unity than  $\text{La}_{0.8}\text{Ca}_{0.2}\text{MnO}_3$  (0.894) and  $\text{Pr}_{0.8}\text{Ca}_{0.2}\text{MnO}_3$  (0.877). Neutron-scattering measurements were carried out using the triple-axis spectrometer TOPAN at the JRR-3M reactor in Japan Atomic Energy Research Institute. Typical condition employed was the fixed final energy at 14.7 meV with horizontal collimation of Blank-30'-Sample-60'-Blank. The (002) reflection of pyrolytic graphite (PG) is used to monochromatize and analyze neutrons. A PG filter was used to reduce higher-order contaminations in the incident beam. The Curie temperature  $T_C$  and its Gaussian distribution  $\Delta T_C$  was determined using the temperature dependence of the  $(100)_{\text{Cubic}}$  peak intensity;  $T_C$  ( $\Delta T_C$ ) is 306(1.1), 281(1.4) and 271(10) K for  $y = 0.00, 0.05$  and  $0.10$ , respectively.

The spin-wave dispersion curves for studied  $(\text{La}_{1-x-y}\text{Y}_y)\text{Sr}_x\text{MnO}_3$  samples show an isotropic behavior in the measured  $(q, \omega)$  range,  $q < 0.40 \text{ \AA}^{-1}$  and  $\hbar\omega < 20$  meV, and follow well with  $\hbar\omega(q) = Dq^2 + E_0$ , typical of ferromagnetic spin-wave dispersion in low  $q$  and low  $\omega$  range. In order to study the temperature dependence of spin-wave stiffness  $D$ , we have performed

constant- $Q$  scans at  $(1.1 \ 1.1 \ 0)$  where well-defined peak profiles are obtained in a wide temperature range. Each peak profile was fitted with the spin-wave scattering cross section including the finite life-time  $\hbar/\Gamma$  convoluted with a proper instrumental resolution. Figure 4 shows thus obtained temperature dependence of  $D$ . Error bars indicate fitting errors. Dashed lines are guides to the eye. Softening of spin wave dispersion  $\omega(q, T)$  is observed as temperature approaches  $T_C$ .

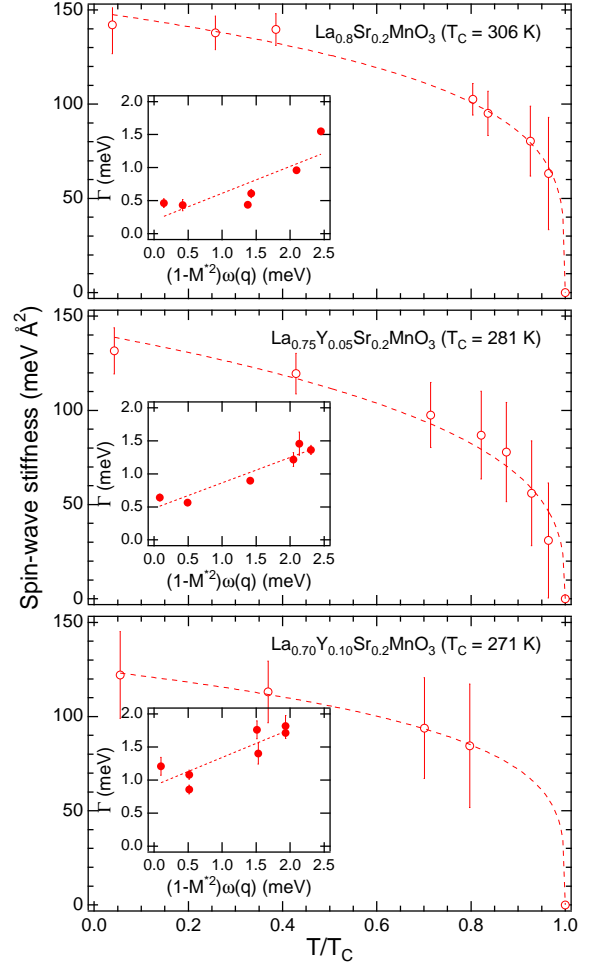


FIG. 4. Experimental results for the temperature dependence of the spin-wave stiffness  $D$ . Error bars indicate fitting errors. Dashed lines are guides to the eye. Inset shows a linear relation between  $\Gamma(q, T)$  and  $(1 - M^{*2})\omega(q, T)$ , where  $M^* = M(T)/M(0)$ .

At the inset of Fig. 4 we show the inverse lifetime  $\Gamma(q, T)$  versus  $(1 - M^{*2})\omega(q, T)$ . From the figure we see that inverse lifetime is fitted in the form

$$\Gamma(q, T) = \Gamma_0(q) + \alpha_q(1 - M^{*2})\omega(q, T). \quad (2.6)$$

Temperature dependence is scaled in a way consistent with theoretical result for DE systems.  $\Gamma_0$  is the  $T$ - and  $\omega$ -independent part of the spin wave lifetime, *i.e.*

$\Gamma(q, T \rightarrow 0) = \Gamma_0(q)$ , which increases systematically by increasing the ratio of Y atoms. We speculate that the origin of  $\Gamma_0$  is mainly extrinsic effects such as disorder and inhomogeneity of the sample.

### III. DISCUSSIONS AND SUMMARY

This result is explained as follows. At  $T = 0$  where local spin moment is saturated, we have so called half-metallic state for itinerant electrons. The Stoner excitation, which is a particle-hole process of different species of spins, lies at high energy part  $\omega \sim 2J_H$ . Then the spin wave excitation spectrum, can be observed without being hidden by Stoner continuum. This is the reason for observation of spin wave collective mode throughout the Brillouin zone in the low temperature regime even though the system is metallic. However, at finite temperature with unsaturated spin moment, the density of states splits into two subbands [2] and there appear low energy Stoner processes which damp the spin wave modes. In Fig. 5 we schematically illustrate the Stoner processes at the ground state and at finite temperature.

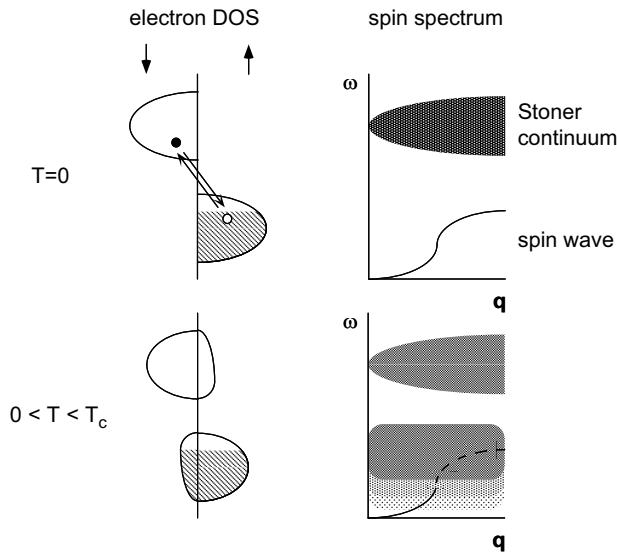


FIG. 5. Electron density of states and the Stoner excitation processes of the DE systems at the ground state and at finite temperature.

Low energy part of the Stoner excitation is produced by the process of occupied electron excited into unoccupied state with opposite spin species. By counting density of states available for the process we see  $\mathcal{I}m\chi \propto \omega$  at  $T \ll W$ . As for temperature dependence, density of state change by temperature in the way that the majority band has the weight  $(1 + M^*)/2$  and the minority band  $(1 - M^*)/2$ , which is the weight for the initial and final state of Stoner processes, respectively. Thus the Stoner absorption is scaled in the form  $\mathcal{I}m\chi \propto (1 - M^{*2})\omega$ .

In the presence of strong electron-lattice coupling such as Jahn-Teller mode, there should exist strong

magnon damping processes. Presence of such strong damping is inconsistent with the experimental fact for (La,Pb)MnO<sub>3</sub> that spin wave excitation spectrum is well-defined throughout the Brillouin zone [18]. Strong damping effect also cause the deviation from the scaling form (2.5). Thus, as long as spin wave excitation spectrum in relatively wide band compounds are concerned, electron-lattice couplings does not seem to be so strong as to affect magnon damping effects.

To summarize, we studied the spin wave lifetime of the CMR systems, theoretically by dynamical mean-field approach for the DE model and experimentally by neutron inelastic scattering of ((La,Y),Sr)MnO<sub>3</sub>. Scaling relations in the spin wave lifetime of DE model as well as CMR manganites is consistent with each other. Spin excitation spectrum of CMR manganites with relatively wide bandwidth is explained by the DE model.

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